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SILVER ISOTOPIC ANOMALIES IN IRON METEORITES: COSMIC-RAY PRODUCTION AND OTHER POSSIBLE SOURCES

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ABSTRACT

The sources of excess 107Ag observed in iron meteorites by Kaiser, Kelly, and Wasserburg (1980) are examined, with emphasis on the reactions of cosmic-ray particles with palladium. The cross sections for the production of the silver isotopes from palladium by energetic cosmic-ray particles are evaluated or estimated and used to calculate spallogenic production rates relative to that of $^{53}\mathrm{Mn}$ from iron. The upper limit for the production rate of excess 107 Ag by galactic-cosmic-ray particles is 400 atoms/min/kg(Pd) which, over an exposure age of 10^9 vears, would make only 1% of the observed excesses of 107 Ag. Neutron-capture reactions with Pd isotopes produce mainly ¹⁰⁹Ag. Binary fission of a siderophilic superheavy element would be expected to yield more 109 Ag than 107 Ag. An intense proton irradiation in the early solar system probably would produce a lower ratio of $(^{107}\text{Pd}/^{108}\text{Pd})$ to $(^{26}\text{Al}/^{27}\text{Al})$ than observed in meteorites. Therefore the presence of excess 107 Ay in iron meteorites with large Pd/Ag ratios very likely is due to the incorporation of 6.5 \times 10^6 -year 107 Pd of nucleosynthetic origin in these meteorites.

INTRODUCTION

A recent addition to the ever-growing list of isotopic anomalies in meteorites is 107 Ag. Kelly and Wasserburg (1978), hereafter denoted KW78, reported excess 107 Ag in samples of the Santa Clara iron meteorite which had large Pd/Ag ratios ($\sim 10^4$). They found that the Pd isotopic ratios in one of their Santa Clara samples were normal and that a sample of Canyon Diablo with a low Pd/Ag ratio had a normal silver isotope ratio. Kelly and Wasserburg (1979) [KW79] gave more results for samples of Santa Clara and reported that Tlacotepec, which is a group IVB ataxite like Santa Clara, had a large Pd/Ag ratio. Kaiser, Kelly, and Wasserburg (1980) [KKW80] found that much of the silver previously analyzed by KW78 and KW79 was contamination. Central pieces of Santa Clara and Piñon (an anomalous iron meteorite) had very large 107 Au/ 109 Ag ratios (up to 2.8 compared to a normal ratio of 1.08). A central piece of Tlacotepec had a normal silver isotopic ratio. The ratio of excess $^{107}\mathrm{Ag}$ to 108 Pd in the Santa Clara and Piñon meteorites were about 1.4 \times 10 $^{-5}$ and 1.0×10^{-5} , respectively (KKW80). The excesses of 107 Ay were considered by KW78 and KW79 as evidence for the existence of 6.5×10^6 -year 107 Pd in the early solar system. However, KKW80 felt that it was important to consider other possible sources of excess 107 Ag and, possibly, of all the silver found in iron meteorites.

Besides late stage nucleosynthesis, the possible sources of silver isotopes given by KKW80 were an intense local proton irradiation in the early solar system, fission of a transuranic or superheavy siderophilic element, and reactions of galactic-cosmic-ray particles with palludium. Lee (1978) tried to explain 16 0 and 26 Al isotopic anomalies as the result of the intense proton irradiation of a small part of the solar system with a proton spectrum which varied with energy as $\mathbb{E}^{-4.5}$. Reeves (1978) argues that lithium isotopes which

would be produced by low-energy alpha-particles reactions in such an irradiation are not found and that an early intense irradiation could not produce the amount of ²⁶Al needed to explain observed ²⁶Mg isotopic anomalies. Libby, Libby, and Runcorn (1979) have proposed that many trace elements in iron meteorites are produced by the fission of a siderophilic superheavy element. The "normal" (within 0.05%) Pd isotopic ratios in a Santa Clara sample are evidence against such a fission source for a significant fraction of the elements in this region of the periodic table which are found in iron meteorites.

Most iron meteorites have been exposed to cosmic rays for periods of 10^8 to 10^9 years; the Piñon and Tlacopetec iron meteorites have measured exposure agas of 790 ± 50 and 345 ± 55 Ma (1 Ma = 10^6 years), respectively (Voshage and Feldmann, 1979). Over such a long exposure, it is possible for the production of a large number of stable nuclides by cosmic-ray-induced nuclear reactions. In iron meteorites, the main target for the production of silver isotopes by cosmic-ray particles is palladium. The reactions with palladium which produce silver isotopes are induced mainly by low-energy secondary neutrons made by the very energetic primary galactic-cosmic-ray particles. Such reactions can produce nuclei in relatively large yields. Detailed calculations of the production rates of silver isotopes in iron meteorites by cosmic-ray reactions are presented here. The other possible sources of the $\frac{10^7}{4}$ Ag isotopic anomalies are also examined.

COSMIC-RAY-PRODUCED NUCLIDES IN METEORITES

The details of the interactions of cosmic-ray nuclei with matter and of the production of "cosmogenic" (cosmic-ray-produced) nuclei have been discussed elsewhere (e.g., Arnold, Honda, and Lal, 1961; Kohman and Bender, 1967; Honda and Arnold, 1967; Reedy and Arnold, 1972). Solar cosmic rays (SCR) are particles

which originate at the sun and which usually have energies below 100 MeV. The galactic cosmic rays (GCR) come from outside the solar system with energies of the order of a GeV. Both types of cosmic rays consist mainly of protons and alpha particles (with a proton/alpha-particle ratio of about 10). Nuclide production by SCR particles in meteorites usually is unimportant because of meteorite ablation in the Earth's atmosphere and of the relatively row fluxes of SCR particles beyond 1 AU. The production of silver isotopes by SCR particles will not be considered here.

The GCR particles produce many secondary particles when they interact with matter. The most important secondary particles for nuclide production are the neutrons (see, e.g., Arnold et al., 1961). In the moon's surface, there are about 13 neutrons/cm² s produced with energies below 10 MeV (Woolum et al., 1975). For meteorites, the incident flux of GCP particles is omnidirectional instead of 2π steradians for a lunar sample. Iron probably yields slightly more neutrons per interaction of a primary GCR particle, but there are fewer iron nuclei per unit thickness (in g/cm²) than in the moon, so the number of secondary neutrons produced in iron meteorites is about twice that in the lunar surface, about 26 neutrons/cm² s for energies below 10 MeV. Approximately 10-15 neutrons/cm² s are produced with energies above 10 MeV (mainly with energies between 10 and 50 MeV) which never slow to energies below 10 MeV. Most of these particles only traverse a small fraction of a meteorite, so actual fluxes at one location are much lower. The fluxes of primary and secondary GCR particles at a 50 g/cm² depth in the moon (Reedy and Arnold, 1972) and in the center of the St. Séverin chondrite (Reedy et al., 1979) are shown in Fig. 1.

Nuclide-producing reactions involving GCR particles roughly can be divided into three categories. Neutrons slowed to energies of KeV or eV produce

nuclides by neutron-capture, usually (n,γ) , reactions. The other two types of GCR reactions involved more energetic particles (E above about 1 MeV) and the emission of one or more nucleons, and are called spallation reactions and the products often are referred to as "spallogenic" nuclides. The two types of spallation reactions involve either "high energy" (E ~ 100 MeV or more) primary or secondary particles or "low energy" (E ~ 50 MeV or less) secondary particles, mainly neutrons. High-energy spallation reactions produce many different nuclides, each one in relatively low yields. Neutron-capture reactions can produce very high yields of a product nuclide if the capturing nuclei has a large (n,γ) cross section (e.g., 158 Gd made by neutron-capture reactions with 157 Gd) and if the parent body is large enough (~100 g/cm² or more) to slow neutrons to energies of the order of eV (see, e.g., Eberhardt et al., 1963). Usually the largest production rates for cosmogenic nuclei involve low-energy, like (n,2n), reactions because both the fluxes of particles and the reaction cross sections are relatively large.

GCR PRODUCTION OF SILVER FROM PALLADIUM

The main target element for production of cosmogenic silver in iron meteorites is palladium. Yields of silver isotopes by GCR-particle reactions with other elements are low because of the low abundances of the target elements, e.g., Cd and In (abundances from the compilation of Mason, 1971), or the large mass differences between the target and product nuclei (e.g., Sn) which means only high-energy reactions can produce the product (with low production rates). The reactions which produce most of the cosmogenic 107 Ag (or 107 Pd) are 106 Pd(n,y), 108 Pd(n,2n), 108 Pd(p,pn), 108 Pd(p,2n), 110 Pd(n,4n) 110 Pd(p,p3n), and 110 Pd(p,4n). The 108 Pd(n,y), 110 Pd(n,2n), 110 Pd(p,pn), and 110 Pd(p,2n) reactions produce 109 Ag or 109 Pd (which rapidly decays to 109 Ag).

The cross sections as a function of energy for the production of 109 Ag and 109 Pd and of 107 Ag or 107 Pd from 110 Pd are shown in Fig. 1. At low energies, the incident particle is a neutron, and, for energies ~100 MeV or greater, the incident projectile is a proton. The 110 Pd(n,2n) 109 Pd cross sections between 10 and 20 MeV were evaluated from the measured and theoretical cross sections of Bormann et al. (1970) and Augustyniak et al. (1977). Be ow 10 MeV and from 20 to 30 MeV, the data of Bayhurst et al. (1975) for 107 Ag(n,2n) and similar reactions were used to get the shape of the 110 Pd(n,2n) excitation function. Above 100 MeV, the cross sections are based on measured (p,pn) plus (p,2n) cross sections with other nuclei. The peak cross section for the 110 Pd(n,2n) 109 Pd reaction is 1.6 barns at 15 MeV. From the trends measured below 28 MeV by Bayhurst et al. (1975), the 110 Pd(n,4n) 107 Pd reaction is estimated to have a peak cross section of about 0.8 b at 36 MeV. The cross sections at high energies for the production of mass-107 nuclei from 110 Pd are estimated from cross sections measured for similar reactions.

Mainly because of the long half-life of 107 Pd (6.5 Ma), there are no measured cross sections for the 108 Pd(n,2n)reaction. At 15 MeV, the total non-elastic reaction cross section of neutrons is about 1.5 b. The other reactions possible with 15 MeV neutrons, mainly (n,p), (n,np), and (n, α), have low cross sections (total of ~15 mb, see Garber and Kinsey, 1976). Thus it seems safe to assume that 108 Pd, like other nuclei in this mass region, has (n,2n) cross sections very close to the total reaction cross sections for energies up to the threshold energy of the (n,3n) reaction. The 108 Pd(n,2n) 107 Pd cross sections used here are essentially those used for the 110 Pd(n,2n) 109 Pd reaction, but with the energies shifted to reflect different threshold energies and the peak cross section lowered slightly (to 1.5 b around 16 MeV) because of the fewer neutrons in the 108 Pd nucleus. Above 35 MeV, the same cross sections as used for the production of mass-109 nuclei from 110 Pd were used.

The cross sections for forming 107 Pd and 109 Pd via (n,2n) reactions are well determined (about $\pm 10\%$) for the energies at which most of the reactions occur, below 30 MeV. The 110 Pd(n,4n) 107 Pd cross sections probably have uncertainties of around \pm 25%, but only 15% of spallogenic 107 Ag is made from 110 Pd (the majority coming from 108 Pd). At the energies involved, 10 MeV or greater, it is very unlikely that there are resonances which can produce spallogenic Ag with much larger cross sections from 108 Pd or 110 Pd than the cross sections used here.

Models for the fluxes of GCR particles have been developed by Arnold et al. (1961) and Reedy et al. (1979) for meteorites and by Reedy and Arnold (1972) for the moon. These models generally predict relative trends for spallogenic nuclei better than they calculate absolute production rates. Therefore production rates of 107 Ag and 109 Ag from Pd are calculated relative to that of 53 Mn from iron. Activities of 53 Mn have been measured in many meteorites and it is made from Fe by "low-energy" reactions. The cross sections used for the production of 53 Mn from iron are those of Reedy and Arnold (1972) below 35 MeV and the experimental proton-induced cross sections of Gensho et al. (1979) above 45 MeV. The measured 53 Mn activities in extraterrestrial matter is about 1.4 times those calculated using these 53 Mn production cross sections (see, e.g., Reedy and Arnold, 1972). This 53 Mn normalization factor of 1.4 will be ignored below because the production ratios 107 Ag/ 53 Mn and 109 Ag/ 53 Mn are calculated and we want upper limits for production of silver isotopes.

The fluxes used to get production-rate ratios were the S(100,E) and S(10,E) spectra of Arnold <u>et al.</u> (1961), the ones for the surface and center of the St. Séverin chondrite of Reedy <u>et al.</u> (1979) and the lunar ones for depths of 0, 50, 100, 200, and 500 g/cm² of Reedy and Arnold (1972). The

extremes in spectral shape and therefore in production ratios are for the 0 and 500 g/cm² depths in the moon. The average calculated production-rate ratios and their standard deviations for these nine spectra, plus the extreme values (in parentheses), for 107 Ag (and 107 Pd) or 109 Ag (and 109 Pd) per unit mass of Pd relative to 53 Mn per unit mass of Fe are 1.15 ± 0.31 (0.69-1.54) and 0.45 ± 0.13 (0.25-0.61). From Pd, the production rate of 107 Ag is 2.6 ± 0.1 times that for 109 Ag. The ratio for 107 Ag production from 108 Pd to that from 110 Pd is 5.8 ± 0.9. To illustrate why a low-energy product from iron like 53 Mn is needed to compare with Ag production from Pd, 107 Ag/ 36 Cl production ratios were calculated and found to range from 9 to 212.

A number of measured 53 Mn activities in iron meteorites are compiled in Kohman and Bender (1967) and Arnold and Honda (1967). The values range from undetectable values (less than 15) to 570 ± 60 dpm/kg(Fe). The average 53 Mn activity was around 300 dpm/kg(Fe). In stone meteorites, Englert and Herr (1978) reported 53 Mn activities of 43 to 557 dpm/kg(Fe) and gave an average value of 478 \pm 84 dpm/kg (Fe). Adopting an upper limit of 600 atoms/min/kg (Fe) for the 53 Mn production rate, the maximum production rates for spallogenic 107 Ag and 109 Ag are 690 and 270 atoms/min/kg(Pd), respectively. Actual silver production rates in most iron meteorites by spallation reactions are probably about 0.4 of these upper limit rates.

The upper limit for the spallogenic production rate of excess 107 Ag is 690 - 270 x (0.5183/0.4817) = 400 atoms/min/kg(Pd), where 0.5183 and 0.4817 are atom abundances of 107 Ag and 109 Ag in natural silver, respectively. Because there are 1.511 x 1024 atoms of 108 Pd per kg of Pd, this production rate of excess 107 Ag is 4.4 x 1024 atoms/s/atom(108 Pd). Most iron meteorites have exposure ages less than 1 Ga (= 3.2 x 1016 s). Assuming an exposure age of 1 Ga, the upper limit for the ratio of excess spallogenic 107 Ay to 108 Pd is

 1.4×10^{-7} . This is about 1% of the observed excesses in the Santa Clara and Piñon iron meteorites.

The only other source mechanism for cosmogenic silver is neutron capture in mass 106 and 108 nuclei. Because the cadmium abundances in iron meteorites are very low, similar to thuse of silver (see compilations in Mason, 1971) and because the atom abundances and capture cross sections (Lederer et al., 1978) for 106 Cd and 108 Cd are only 1.25% and 1 ± 1 b and 0.85% and 1.2 ± 0.36 b, respectively, the production of silver isotopes by neutron-capture reactions with cadmium should be negligible. The atom abundances and capture cross sections for 106 Pd and 108 Pd are 27.3% and 0.29 ± 0.03 b and 26.7% and 11 ± 2 b, respectively (Lederer et al., 1978). Thus 109 Ag is produced 37 times faster than 107 Ag by thermal neutron-capture reactions with Pd. The production ratio using resonance integrals is 109 Ag/ 107 Ag = 43. Therefore an excess of 107 Ag can not be produced by neutron-capture reactions, and neutron-capture reactions would diminish the amount of excess 107 Ag made by spallation reactions.

Rates for neutron-capture reactions in iron meteorites are not well determined and can vary considerably with the pre-atmospheric size of the meteorite (Eberhardt et al., 1963; Van Dilla et al., 1960). The radionuclide 60 Co is produced mainly by the 59 Co(n, γ) reaction. Activities of 60 Co in the Yardymly (often called Aroos) iron meteorite (which had a very high 53 Mn activity of 570 ± 60 dpm/kg) and in the Sikhote-Alin iron meteorite (which was very large) were 17 ± 2 and 95 ± 10 dpm/kg, respectively (Honda and Arnold, 1967). These activities correspond to "thermal" neutron fluxes of about 0.15 and 0.83 neutrons/cm² s, respectively. These fluxes would give rates for the 108 Pd(n, γ) reaction of about 150 and 836 atoms/min/kg(Pd), respectively. The inclusion of epithermal (resonance) neutrons in considering neutron-capture production of 109 Ag probably would increase these rates. Thus some, or in

large iron meteorites, even all of the spallogenic excess 107 Ag could be "cancelled" by neutron-capture-produced 109 Ag. Iron has a low total cross section for neutrons with energies near 24 keV (see compendium of Garber and Kinsey, 1976), so elements with resonances in this energy region could have enhanced neutron-capture rates in iron meteorites. However, Pd and Cd do not have any strong (n,γ) resonances in this energy region (Garber and Kinsey, 1976). In summary, the upper limit for the production rate of excess 107 Ag by cosmic-ray reactions with Pd in iron meteorites is 400 atoms/min/kg(Pd), and the actual rate of making excess 107 Ag in most iron meteorites would be quite smaller.

DISCUSSION

The excess 107 Ag (denoted hereafter as 107 Ag*) found in the Santa Clara and Piñon iron meteorites can not be made by reactions of cosmic-ray particles with Pd, as the measured 107 Ag*/ 108 Pd ratios are 1.0×10^{-5} and 1.4×10^{-5} , respectively (KKW80), and the upper limit for cosmogenic 107 Ag* produced during a 10^9 year exposure is 107 Ag*/ 108 Pd < 1.7×10^{-7} . The upper limit for 107 Ag*/ 108 Pd, 3 x 10^{-6} , measured in Tlacotepec, which had an exposure age of 945 ± 55 Ma, is consistent with this upper limit for cosmogenic 107 Ag*. For reasons given above, the actual cosmogenic 107 Ag*/ 108 Pd ratio in most iron meteorites is probably around 5 x 10^{-8} or lower.

As mentioned above, other possible sources for 107 Ag* were nucleosynthesis, fission of a siderophilic superheavy element (see Libby et al., 1979), and an intense local proton irradiation in the early solar system (Lee, 1978). The essentially normal Pd isotopic ratios measured by KW78 for a Santa Clara sample does not exclude the possibility that some excess 107 Ag is due to the fission of a superheavy element (SHE). The fission of a nucleus with 2 107 Ag

produces more 107 Ag than 109 Ag because such nuclei fission asymmetrically with fission-product yield peaks near A = 90-105 and A = 139 and an intermediate valley of low yield (see, e.g., Hoffman and Hoffman, 1974), and such a fission pattern produces more mass-107 than mass-109 nuclei. However, fermium (Z = 100) nuclei fission symmetrically or with the 'ow-mass peak above A = 110 (Hoffman and Hoffman, 1974), so the 107/109 yield ratio is less than 1. Elements with Z > 100 probably fission in a manner similar to Fm, so it seems unlikely that fission of a SHE or any trans-fermium nuclide would produce an excess of 107 Ag. The absence of fission products with A < 110 might not be good reidence against the presence of SHE fission products in an iron meteorite as the lowest yield peak for SHE binary fission might be A ~ 140 and thus yields of A < 110 nuclei would be quite low. Also, the fission mechanism of SHE nuclei could be quite different from the binary fission processes observed for nuclei with Z < 100.

Lee (1978) proposed that an early intense local proton irradiation with protons having an energy spectrum of E $^{-4.5}$ produced 16 O and 26 Mg isotopic anomalies. The excess 26 Mg in some meteorites is ascribed to material having 0.72-Ma 26 Al and 27 Al with a 26 Al/ 27 Al ratio of 5 x 10 $^{-5}$ (Lee, 1978). The excess 107 Ag in the two iron meteorites due to the decay of 6.5-Ma 107 Pd required a 107 Pd/ 108 Pd ratio of 1.0-1.4 x 10 $^{-5}$. The low-energy nuclear reactions which could produce 107 Pd are 106 Pd(n, γ), 107 Ag(n, ρ), and 108 Pd (p.pn). Only the last reaction is induced by low-energy protons. Cross sections for the 108 Pd(p,pn) 107 Pd reaction were estimated from analogous reactions and with an assumed peak cross section of 0.25 b around 25 MeV. The cross sections of Reedy and Arnold (1972) were used for the 26 Mg(p,n) 26 Al reaction (similar to those used by Lee, 1978). The production-rate ratio in a E $^{-4.5}$ proton spectrum of the 26 Mg(p,n) 26 Al and 108 Pd(p,pn) 107 Pd reactions was 9. Assuming a 26 Mg. 27 Al

ratio of 1.3 (from hondritic values in Mason, 1971), ²⁶Al relative to ²⁷Al is produced 12 times faster than ^{107}Pd from ^{108}Pd . The observed ^{26}Mg and ^{107}Ag anomalies imply a relative production ratio of around 4. One way to get this ratio of 4 from the ratio of 12 calculated for the $E^{-4.5}$ spectrum is that the proton irradiation be stopped and ²⁶Al allowed to decay for about 1.2 Ma before condensation. A harder proton spectrum (lower absolute value of the power-law spectrum exponent) would increase the rate of the ¹⁰⁸Pd(p,pn)¹⁰⁷Pd reaction relative to the 26 Mg(p,n) 26 Al reaction rate, but the production of ²⁶Al by the ²⁷Al(p,pn) ²⁶Al and ²⁸Si(p,n2p) ²⁶Al reactions would also increase, raising the $(^{26}AI/^{27}AI)/(^{107}Pd/^{108}Pd)$ production ratio. A harder spectrum probably would mean that other possible isotopic anomalies which are not observed would have higher production rates. If most of the irradiated matter were in grains of $\sim 0.1 \text{ g/cm}^2$ radii, the lowest energy protons would be stopped and the (p.pn) products would have a higher yield relative to a (p.n) product (see SCR fluxes and production profiles of knedy and Arnold, 1972). While an early intense proton irradiation can not be ruled out as producing ²⁶Al and 107 Pd in the early solar system, the 107 Pd/ 108 Pd ratio being near that of 26 Al/ 27 Al makes the parameters of such an irradiation more complex and makes it less likely as a source mechanism of the $^{26}\mathrm{Mg}$ and $^{107}\mathrm{Ag}$ isotopic anomalies.

The excess 107 Ag which was observed in the Santa Clara and Piñon iron meteorites can not be produced by a long exposure to cosmic-ray particles and its production by fissic seems very unlikely. Therefore the titles of the papers by KW78 and KW79 were correct, that is, there is good evidence for the existence of 107 Pd in the early solar system. It is harder to make the amount of 107 Pd observed in these iron meteorites by an early intense proton irradiation than it is to make the 26 Al seen in other meteorites by such a mechanism. Therefore the excess 107 Ag is very likely due to the decay of nucleosynthetic 107 Pd in these iron meteorites.

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FIGURI CAPTION

Fig. 1. Cross sections (in barns on left axis) for the GCR production of ^{109}Pd and ^{109}Ag (solid triangles) or of ^{107}Pd and ^{107}Ag (solid circles) from ^{110}Pd , plus the GCR particle fluxes (on right axis) of Reedy et al. (1979) for the center of the St. Séverin chondrite (dotted line) and of Reedy and Arnold (1972) for a 50 g/cm² depth in the moon (solid line) are shown as a function of particle energy. The scale for the energy axis changes at 100 MeV.

